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Final Report

Contract Number	ONR N00014-99-1-1087				
Title of Research	New Techniques in Photo-Acoustic Spectroscopy				
Principal Investigator	T.F. Morse				
Organization	Laboratory for Lightwave Technology, ECE, Boston University				

I. Technical Objectives

The technical objectives of this effort are to develop tunable fiber lasers that may be amplified by double clad fibers. We have demonstrated that it is possible to obtain continuous tuning of an erbium fiber laser between 1527 nm and 1572 nm. We have also been able to tune a Tm laser between 1700 nm and 2000 nm. By mixing these two wavelengths in a non linear crystal, it should be possible to obtain continuous tuning of radiation from 6.5-20 microns. In order for the non-linear interaction in the crystal to be efficient, it is necessary that the radiation consist of intense, polarized pulses. The goal of this work is to provide a tunable, pulsed source that is in the 7-20 micron range. It should also be compact and relatively inexpensive. This would provide pulsed radiation that can be used in conjunction with photo-acoustic spectroscopy for the detection of chemical agents with absorption signatures in this wavelength region.

II. Technical Approach

1. Basic Concept

The fundamental concept behind this work is to put together various components as building blocks that will ultimately result in a source of tunable high peak power pulsed radiation in the infrared (7-20 micron) region of the spectrum. The schematic of this concept is illustrated in Figure 1. The components consist of two tunable pulsed sources, a means of Q switching them, and a double-clad amplifier fiber to increase the intensity of these pulses. The pulses are then mixed in a nonlinear crystal to produce radiation at the frequency differences of the two pulses. We note that all of these elements have been reported on separately, and in this work our intention was to combine them in a novel manner. We shall consider each of these aspects in turn.

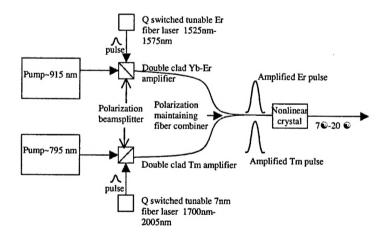


Figure 1. Schematic of Tunable Pulsed IR Laser

2. Aerosol Deposition for Fiber Lasers

The fiber lasers under consideration here are doped with Yb-Er and Tm. The erbium fiber (in the C band alone), exhibits gain from 1,527 nm to 1572 nm. The Tm fiber has gain over an even wider wavelength range, from 1,700 nm to 2,009 nm. We have developed a technique using organometallic aerosol precursors in MCVD (Modified Chemical Vapor Deposition) t fabricate these fiber lasers. Since fiber lasers with arbitrary rare earth doping are not readily available, we consider the fabrication of specialty fibers an important part of this effort. Most fiber lasers, as a consequence of the fact that the rare earth precursors have low vapor pressures, are made with solution doping. In this process, an unsintered core frit in MCVD is soaked with a nitrate or chloride solution of the desired rare earth. This is then converted to the oxide, dried, sintered, and collapsed to provide the rare earth doped preform that is subsequently pulled into fiber. We have proposed an alternate route that we believe to be a better technique for the fabrication of fibers to be doped with materials that have low vapor pressure precursors. This includes all of the rare earth elements. In this process, we use a solution of organometallic compounds, with tetra-ethyl ortho siloxane (TEOS) as a solvent for the rare earth beta-diketones (solids). The mixture containing the dissolved solids is then nebulized using a 1.6 MHz transducer. The resulting aerosol particles are of the order of a few microns. The size is such that the dissolved solids may be convectively transported in MCVD. [Morse, et al., 1989, 1991, 1993] The aerosol is depicted in Figure 2.

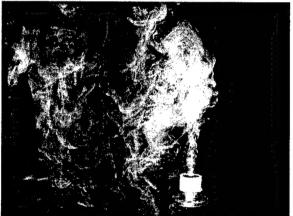


Figure 2. Aerosol of Organometallic Precursors

The precursors are similar to those used in a sol gel process in that the reactants and reactant products are similar; however, the reaction pathways for these two processes are significantly different. A sol gel reaction occurs usually at room temperature, with a slow cross linking to provide the silica matrix. Using organometallics in MCVD, the temperature is over 1,000 C. Consequently, the reaction occurs in milliseconds rather than hours. The precursors "explode" (vigorously decompose), and the molecular fragments provide the basis of a gas phase homogeneous chemical reaction that provides the multi component oxide for the preform core. This is a new way of making glass. In Figure 3 we show a schematic of the use of the aerosol technique in MCVD with a Pt sting to provide electrophoretically assisted deposition.

An indication of the efficiency of the above described aerosol process is illustrated in Figure 4. This depicts the losses of an erbium doped fiber laser, with

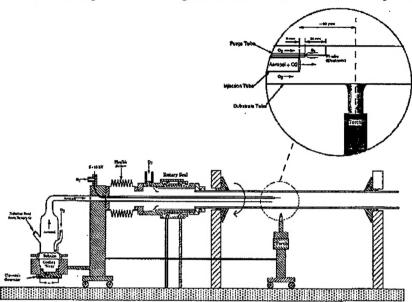


Figure 3. Electrically assisted aerosol deposition in MCVD

a relatively high erbium content. Note the low water peak at 1,380 nm and the loss minimum at 1,080 nm that indicate the effectiveness of the aerosol technique.

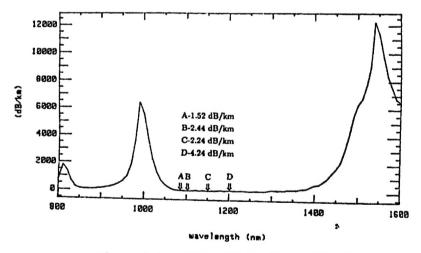


Figure 4. Low loss erbium doped silica fiber laser

3. Tuning Ytterbium/Erbium and Thulium fiber lasers

We realize the necessity of the use of polarization control in the non-linear mixing of two wavelengths in a non-linear or quasi-phase matched crystal, and we are able to fabricate fiber lasers so that they will have a single polarization. This can be

accomplished with the traditional Panda design, indicated in Figure 5. Polarization maintaining fiber is obtained by drilling small holes in the preform in which polished rods of a different thermal expansion coefficient are inserted. In this manner, a highly anisotropic stress distribution is induced when the preform is pulled into fiber. This was to have been done for the fiber used for the "seed" signal as well as for the double clad amplifier fibers. Polarization maintaining double clad fibers, we believe, are not presently commercially available. We did not proceed far enough in this project to accomplish this.

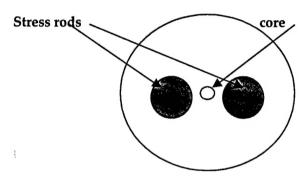


Figure 5. Panda Design for Polarization Maintaining Fiber

In addition, in order for the non-linear frequency mixing to be efficient, it is also necessary that the laser radiation be single mode (transverse) and tunable. Since we desire a broad tuning range, the wavelength narrowing aspects of a Bragg grating in the fiber can not be used. However, with a pumping scheme employing a directional coupler [Scrivener, 1989], tunable laser radiation as narrow as 1.0 Å has been achieved. This value was limited by the resolution of the spectrometer measuring the linewidth. Effective linewidth is also determined by the number of longitudinal modes, and this is determined by the cavity length, or whether mode selective elements have been placed within the cavity. In a ring configuration with the addition of internal loops, it has been demonstrated that single frequency lasing can result. [Urquart, 1988] This technique is compatible with one we have developed for broad band tuning of optical fiber lasers that will be described in the following. [Kozlov, et al., 1998, Shubochkin, et al., 1998]

In order for the proposed technique to be viable in the creation of frequency differenced pulses, we must be able to tune the laser over the gain bandwidth of the fiber laser. There are various techniques for tuning fiber lasers. A bulk intra-cavity dispersive grating can be used. This can provide a wide tuning range; however, there are disadvantages in using a bulk element (increased intra-cavity loss). Some previous efforts (Reekie, et al, 1986) have indicated that the tuning is not continuous across the erbium gain bandwidth. If Bragg gratings are used to configure the laser cavity, it is possible, through strain tuning of these elements, to change the lasing wavelength. In this case, the tuning is limited to the order of perhaps 10-20 nm. Another type of tuning can be obtained by using a polished quartz evanescent wave directional coupler in a ring configuration. This too, has the disadvantage of being difficult to control, since a mechanical displacement of the two polished quartz elements is needed to achieve tuning. In addition, this tuning has proved to be discontinuous. [Scrivener, et al., 1989] Tuning can also be achieved with an acousto-optic modulator. In this scheme, the tuning may not be continuous across the gain bandwidth. [Wysocki, et al, 1990]

An alternate, and novel tuning method that we have developed is employed in which a fused taper 2 x 2 coupler is used in a loop mirror configuration. [Kozlov, et al., 1998, Shubochkin, et al., 1998] It is known that if it were possible to change the reflectivity of an output coupler in a given lasing system, then, as this output coupling, or mirror reflectivity is changed, the laser will shift in wavelength. For the first time, we have accomplished this with a simple 2 x 2 HOCC (Highly OverCoupled Coupler) used as a loop mirror. This loop mirror is used as the output coupler of a fiber laser. A broad band mirror and the loop mirror are used to define the laser cavity. This is schematically illustrated in Figure 6.

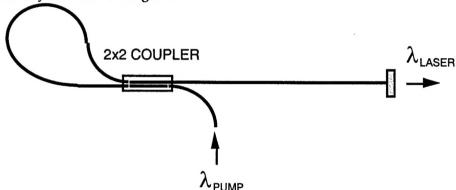


Figure 6. Fiber laser with loop mirror using a 2 x 2 fused taper coupler

The reflectivity of a loop mirror will be periodic in wavelength, and, in order to insure that only one single wavelength lase within the gain curve (there will still be many longitudinal modes within this wavelength0, a 1/4 wave of the periodicity, which is illustrated in Figure 7, should correspond to the gain bandwidth of the fiber laser. Since

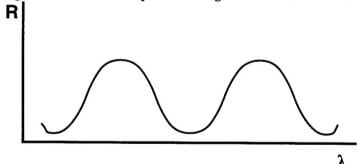


Figure 7. Periodic behavior of 2 x 2 coupler used as a loop mirror

the cavity described in Figure 6 will lase at the maximum reflectivity of the loop mirror, it is this characteristic that insures that there will be only one wavelength lasing across the gain spectrum. If the peak reflectivity of the loop mirror can be changed as a function of wavelength, then we have a means of tuning the fiber laser. The loop mirror reflectivity may be tuned by stretching the waist section of the coupler. As the waist section of the coupler is elongated, the sine wave like reflectivity shifts continuously in wavelength. Straining the narrow waist section of the HOCC can be problematic, since this section is extremely thin. Temperature tuning, however, has proved to be more efficient. The waist section of the coupler is embedded in a UV curable polymer with a

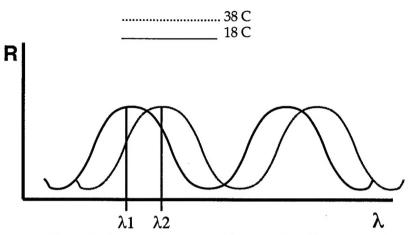


Figure 8. temperature dependence of HOCC (Highly OverCoupled Coupler) Fiber Mirror

high dn/dT. If the coupler is attached to a compact Peltier plate, as the plate temperature changes, the reflectivity of the HOCC loop mirror shifts, and the peak reflectivity shifts to longer wavelength. If this loop mirror is used as the output mirror in a linear erbium or thulium fiber laser, with the pump radiation being transmitted through a dichroic mirror broad band at 1.55 μm for erbium and at 1.9 μm for Tm, then, by heating the loop mirror (effectively changing the refractive index of the cladding material of the waist section of the interaction region of the 2 x 2 coupler) we can tune erbium continuously over the gain bandwidth from 1.527 μm to 1.574 μm .

This can be achieved with a temperature change of 20°C in a linear or in a ring configuration. The same principal applies to the tuning of a Tm doped optical fiber laser. Using a HOCC of differing number of cycles, we have been able to tune a Tm doped fiber laser from 1810 nm to 1930 nm. We are confident that this wavelength tuning range can be extended, although a HOCC with a different number of cycles may be required.

Diode-pumped, broadly tunable, single frequency fiber laser based on compound optical-fiber based resonators have been reported. [Urquart, 1988] Tuning and single longitudinal mode selection were accomplished by use of two fiber Fabry-Perot etalons, [Zyskind, et al, 1991] an all-fiber compound-ring resonator in which a dual-coupler fiber ring is inserted into the main cavity [Zhang, et al., 1996] or with a passive multiple-ring cavity [Lee, et al., 1998]/ To maintain single-mode operation when the wavelength is changed, it was necessary to adjust the polarization controller in the ring cavity to suppress a second mode which appeared to arise from birefringence in the ring. Use of polarization maintaining fiber in the ring should eliminate this source of instability [Hernandez-Cordero, et al., 1998]

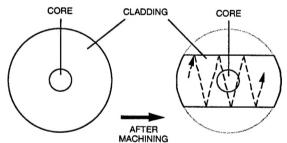
4. Q switched pulses

Thus far, we have shown how it is possible to tune two fiber lasers over a wavelength regime whose difference would result in continuously tunable radiation from 7 μ m to 20 μ m. In order for these photons to "mix" efficiently in a nonlinear crystal, we must insure that the fibers are polarization maintaining. We have the ability to fabricate polarization maintaining rare earth doped fiber; however, preliminary experiments have used non-polarization maintaining fibers

with the polarization controlled using polarization "paddles". The Q switching can be accomplished by several techniques. Our experiments have been carried out using an acousto-optical modulator. This is a "brute force" technique, the bulk components are not inexpensive, and a final "field" instrument would certainly prefer to use a passive technique. Our experiments produced were able to produce pulses that were or the order of 1 microsecond, which is too long for appreciable peak powers, needed to achieve high efficiency in non-linear mixing. Work was in progress to improve this. Saturable absorbers are another possible route to pursue; however, recent work at Southampton University has shown that a metal eutectic thin film mirror that receives a high energy pulse will melt, its reflection characteristics will change (i.e., the reflection will significantly decrease), and that this response is fast enough for efficient Q switching. Should this work be pursued further, this approach shows the most promise.

5. Double clad fibers

One key aspect of this effort has been to develop a reliable means of amplifying a short, tunable fiber laser pulse. This can best be accomplished using a double clad fiber. This concept can best be described in the following. As laser diode power increases, there is a desire to pump the core of single mode fiber lasers. The brightness theorem states that the product of the diode numerical aperture and area from which the diode light emanates must be a constant. Since the single mode core area is quite small, it is not possible to focus the diode light into the core. Thus, highly inefficient pumping results. In order to overcome this drawback, Snitzer, et al., have proposed the following. If we consider a structure such as shown in Figure 9, the light from the diode, or diode array, is focused into the cladding material surround the single mode core.



Typical fiber preform machined to eliminate helical rays

Figure 9. Schematic of Double Clad fiber

This first cladding, in turn, is surrounded by a lower index material, usually a UV curable polymer. Thus, the first cladding serves as a multimode guiding structure for the pump photons. As this radiation travels down the fiber, any photons that traverse the core will be absorbed by the rare earth doped material in this single mode core. The absorption is proportional to the (core area)/(first cladding area). For this reason, such double clad fibers are often of the order of several tens of meters in length to guarantee that all of the pump are absorbed. In this manner, the Brightness Theorem is circumvented, and the double clad fiber functions not only as a wavelength converter, but a modal and brightness converter. The high power pump diodes are multimode and the fiber output is single mode. The brightness increase using this technique can be three orders of magnitude.

If we consider a ray description, then many of the pump photons will behave as helical rays, and these will merely have a trajectory that spirals around the core. In order to avoid the loss of

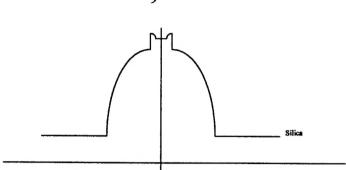


Figure 10. Index profile of all-glass double clad fiber laser

these pump photons, many different fiber designs have been proposed to break the symmetry so that helical rays will be able to be absorbed by the core.

Double clad fibers are usually fabricated using the MCVD process, in which the MCVD substrate tube functions as the first cladding with the doped single mode core inside of this. This means that the outer cladding must have a lower refractive index than silica. To achieve this lower refractive index, the preform is pulled using a low index, UV curable polymer as the second, or outer cladding. With recently obtainable polymers, a numerical aperture of .4 can be readily obtained, which is notably larger than that obtainable from a sleeved tube down doped with fluorine and available from Heraeus. This numerical aperture is approximately 0.2. Although the polymeric outer coating is satisfactory from the standpoint of numerical aperture, if any of the high power pump radiation interacts with the polymer coating (this does not include the evanescent wave), then the double clad fiber is destroyed. An all glass double clad fiber is to be preferred.

We have proposed and have achieved success with the following technique that leads to an all glass double clad fiber. The aerosol deposition technique permits convenient doping with tantalum oxide using tantalum sec butoxide dissolved in TEOS as the precursor. Tantalum oxide modifies silica to increase the refractive index. It has no absorption bands in the infra red and is a good host for rare earth ions. Using our aerosol deposition technique, it is possible to deposit a large multimode inner cladding up-doped from the silica substrate tube refractive index. At the center of this multimode structure is the rare earth doped core with an even higher refractive index. This refractive index profile is illustrated in Figure 9. This can be accomplished with tantalum doping as a consequence of the fact that tantalum oxide has a much smaller thermal expansion mismatch with silica than does germanium dioxide. Achieving such a profile in a germanium doped glass with a large index difference is extremely difficult, since the doped germanium layers to form the multimode first cladding will spall off the inner wall of the silica tube during processing. In addition, we can achieve a high enough numerical aperture (between the first and the second cladding) for this concept to be compatible with a Polychrome Laser from Boston Laser, Inc.

Finally, we note that in order to maintain the polarization of the two pump pulses, the amplifying fiber should also be polarization maintaining. This can be done by the insertion of stress rods in the double clad fiber preform.

6. Amplification of tunable laser pulses

Thus far, we have indicated how a polarization preserving fiber laser may be tuned over the gain bandwidth of the doped fiber, how it may be Q switched, and how a double clad fiber can play a role in the amplification of the Q switched pulse. In this

section, we describe the optical system that permits the convenient amplification of the pulse using a Boston Laser Polychrome diode source. This is shown in Figure 11.

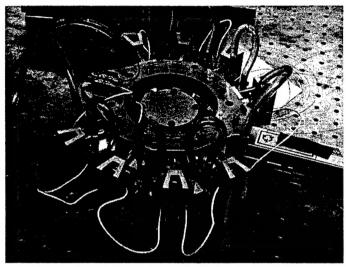


Figure 11. Polychrome Diode Pump Diode Source

In this configuration, eight water cooled diodes focus pump light into a faceted rod that transmits the radiation into a 200 micron, 0.22 NA, step index fiber. The sum of the output power of the eight diodes is 48 W, and the fiber coupled to the diodes is able to transmit slightly over 40 W. This is at 915 nm for pumping an Er-Yb double clad fiber, or at 795 nm, for pumping a double clad Tm fiber. As shown in Figure 11, the optical configuration consists of 8 radially positioned diodes with optics for each individual diode to assure efficient focusing of the light into the fiber. In Figure 12, we see the pattern of diode light as it is focused on the fiber end. It consists of the eight lines from the individual diodes. This light, whether at 915 nm to pump Er-Yb, or at 785 nm, to pump Tm, causes the inversion in the single mode core of the double clad fiber. As noted above, the mixing of two wavelengths in a nonlinear crystal to obtain difference frequencies is dependent upon polarization states. We have previously noted that the

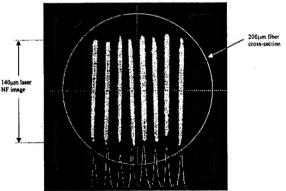


Figure 12. Focused Pattern of Eight Diodes on Fiber End

radiation from our tunable fibers must be polarization maintaining. This permits the use of a polarization beam splitter for combining the seed signal from the tunable fiber and the pump radiation from the high power diode source. This is shown in Figure 13. The efficiency of this process, for high energy pulses, can be of the order of 20%. Success in this undertaking would provide a new, compact instrument for photoacoustic spectroscopy.

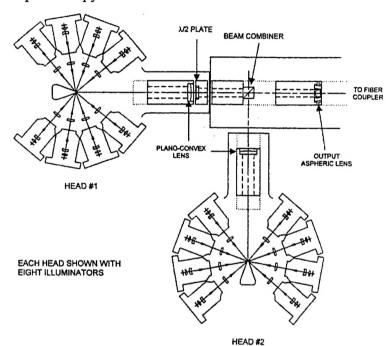


Figure 13. Polarization Combining of Pump and Signal in Double Clad Fiber

This configuration permits combining of the Q switched, tunable, "seed" signal from the fiber with the diode pump photons that prepare the gain core of the double clad fiber. It is possible that proper coiling of the double clad fiber will permit the retention of the polarization properties of the "seed" pulse. If this is not the case, then it will be necessary to make a double clad fiber that is polarization maintaining. This can be done by fabricating a "Panda" preform with appropriate stress rods.

7. Interaction in a non-linear crystal

The focus of this effort has been to attempt to generate high power tunable laser pulses that can be mixed in a nonlinear crystal to generate frequency differences. This generation is a well known technique, and the central focus of this research is to create tunable pulses in the proper wavelength region so that the final output will be continuously tunable radiation from 7 μ m to 20 μ m. The efficiency of this nonlinear conversion can be of the order of 20%.

III. Summary

It is perhaps appropriate at this point to reiterate the basic concept of the proposed effort. By Q switching tunable optical fiber lasers in the 1,527 nm-1,575 nm range (Yb-Er), and 1,700-2,000 nm range (Tm) we can amplify these pulses with a double clad configuration. These pulses can then mix in a nonlinear crystal to produce

frequency difference pulses that can be continuously tuned between 7 $\mu\,m$ to 20 $\mu\,m$. The aerosol technique described above has been implemented in a new configuration on our SCG MCVD system, and this is shown in Figure 13. This system has allowed us, using tantalum sec-butoxide as one of our aerosol precursors, to create a silica glass doped with Ta_2O_5 . This oxide has a large refractive index, it has no absorption bands in the infrared, and, most important, it can serve as a host for rare earth ions. For the first time, we have been able to incorporate over 6,000 ppm (weight) of a rare earth oxide in a silica glass without the use of Al_2O_3 . Lifetimes are such that we believe that concentration quenching is severely reduced which results in higher concentrations, and thus, shorter amplifier sections.

IV. Future Efforts

Our program did not accomplish as much as had been initially anticipated. The fundamental concepts of this proposed effort are, we believe, able to make a significant contribution to the creation of tunable, compact sources in the far infrared where many chemical agents have a signature. The reason, although not the justification, as to why not more was accomplished in this research lies in the following. Our laboratory was moved during the course of this research from Brown University to Boston University, and the setting up of the laboratory took longer than anticipated. In addition, a key member of this effort, Dr. Valery Kozlov, unexpectedly left before the completion of the project. We hope that it will be possible to continue efforts on this program to achieve what we believe might be significant contributions to the creation of tunable sources for photoacoustic spectroscopy.

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